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THEORY OF FORMATION OF NEW KINDS OF SILICATE MATERIALS

ON DATA OF SILICON ORGANIC COMPOUNDS

A. P. Kreshkov

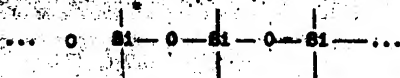
Submitted by Acad D. S. Belyarkin

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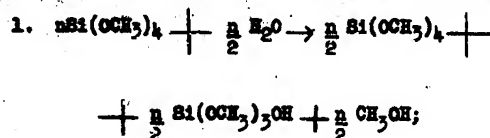
New materials based on the organic compounds of silicon are now being synthesized. They resist the action of corrosive agents, possess excellent mechanical durability and high dielectric constants, are water- and heat-resistant. Consequently, they are widely used in various technical fields.

The manufacture of these materials is based on silicic acid esters and their alkyl and aryl substitution products mixed with the alkoxy-, amino-, and halogen derivatives of silanes, etc.

These compounds can be subjected to saponification, heat transformation, and condensation, thus forming high-molecular silicon organic compounds, characterized by the presence of siloxane groups of silicon and oxygen atoms:



The process of formation of high-molecular silicon organic compounds, for example, from orthosilicic acid esters, may be represented as follows (6):



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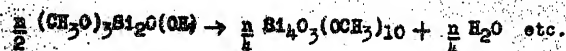
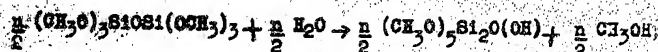
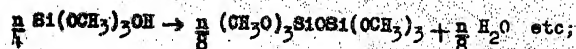
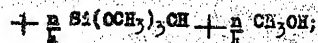
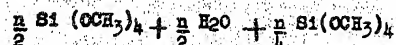
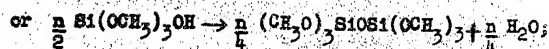
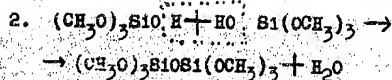
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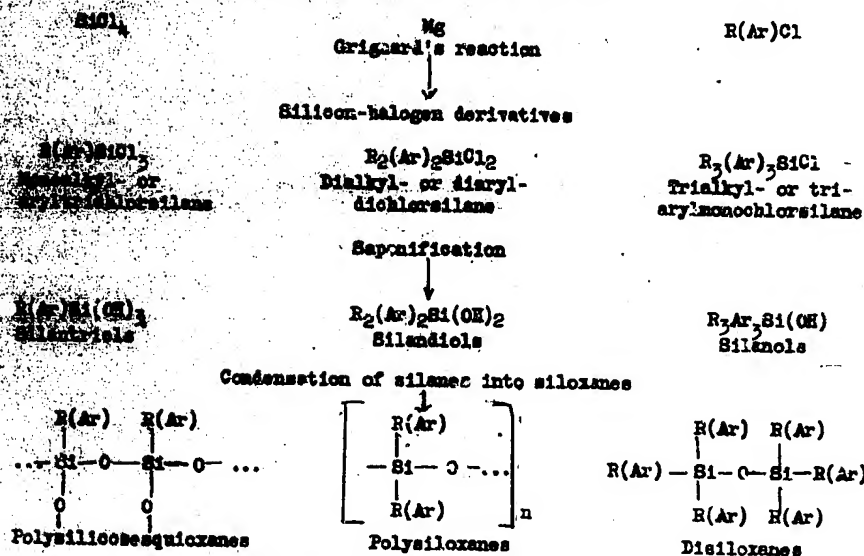
The resultant products are: $(\text{CH}_3\text{O})_3\text{SiO} \cdot \text{Si}(\text{OCH}_3)_2 \cdot \text{Si}(\text{OCH}_3)_3 + 2\text{CH}_3\text{OH}$, i.e., the reaction consists of alternate saponification and condensation.

The relation between the degree of condensation of an ester p and the number of gram-molecules of water n acting on a gram-molecule of the ester is expressed by the following formula:

$$p = \frac{n}{n-1}$$

where n is the number of gram-molecules of the original ester.

Working from alkyl- or aryl-substitution halogen silanes, the formation of polysilicic compounds can be represented as follows:

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In addition to open chain compounds, products whose structure is cyclic are also obtained.

Siloxane chains are very strong. The expenditure of considerable energy is required to break the link between the silicon and oxygen atoms. The energy of combination of oxygen with silicon is 89.3 kilocalories per gram-molecule; the energy of combination of oxygen atoms with each other is 58.6 kilocalories per gram-molecule, which is only $2/3$ of the energy of combination of oxygen with silicon.

The great resistance of silicon organic compounds to the action of heat is due to this. Thus, their exceptional thermal stability, immunity to moisture and many chemical reagents, and excellent dielectric figures are due to the fact that the skeletons of silicon organic compounds are constructed of silicon and oxygen atoms, similar to quartz, glass, and mineral silicates. They are, therefore, synthetic substitutes for fiber glass, asbestos, and mica, compared with which they are excellent artificial materials, resistant to moisture and high temperatures.

However, should the thermal treatment exceed the permissible temperature limit for organic compounds, the organic portion of the high-molecular silicon organic compounds running throughout the highly dispersed basic mineral material undergoes further chemical transformation, connected with an increase in the value of the coefficient of polymerization n .

Therefore, the alternate processes of saponification, condensation, and thermal decomposition at high temperatures ultimately (if the basic material is a nonsubstituted ester) result in the formation of a polymer whose composition approximates $(SiO_2)_n$.

For reasons already given, the physical and chemical properties of high-molecular products obtained from macromolecular silicon organic compounds approximate those of strongly dehydrated polysilicic acids and their salts.

The high-molecular silicon organic compounds thus obtained form a crystalline lattice consisting of molecules forming a chain of elementary nuclei of silicon atoms surrounded by four oxygen atoms disposed at equal distances (1.55 Angstrom Units) from each other on the peaks of an almost perfect tetrahedron.

A lattice frame of this type, formed of silicon and oxygen atoms, resembles the steel structure of a skyscraper, where the intervals between the metal structures are filled with bricks, blocks, tiles, and other building material.

In the construction of ceramic materials of this type, for example, the intervals in the silicon organic shell between the tetrahedral nuclei of silicon and oxygen atoms are filled by other molecules, e.g., by the hexagonal bipyramidal rhombicuboctahedrons of aluminum oxide. As a result, very stable plastic masses, characterized by high mechanical, electrical and other physical properties which enable the finished product to withstand severe working conditions, are formed. The filler, which does not react with silicon organic compounds, is converted at moderate temperatures as a viscous, resilient plastic surrounding the silicon organic polymer.

To this is due the value of the silicon organic compounds used in the manufacture of cement, refractory materials, special ceramics, etc.

Another important use is due to the fact that the following processes occur when high-molecular silicon organic compounds are subjected to the action of heat:

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1. Surface solution of the filler in the binding silicon organic polymer under high-temperature conditions. As a result, a solid solution may be formed, characterized by maximum structural density and high mechanical strength.

2. Formation of chemically active centers around the silicon atoms, characterized by the presence of free units of chemical affinity at the moment the organic radicals are separated from them. This brings about an increase of the tendency for chemical combination with the active molecules of the secondary ingredients, joint crystallization, and the appearance of intensified adsorption.

This suggests the feasibility of using active types of chemical compounds as raw material, capable not only of serving as unusual fillers for the intramolecular spaces of silicon organic polymers or the products of their complete thermal decomposition (silicon-oxygen lattices), but of combining chemically with the active centers of the silicon polymers, when it is desirable to facilitate the acceleration of the reaction of the basic liquid phase (the silicon organic compounds) with the solid phase (metallic oxides, etc.).

The production of high-quality silicate materials from silicon organic compounds envisages the preliminary attainment of high dispersion in the basic material and the employment of guaranteed methods for investigating their dispersion, specific surfaces of the particles, adsorption properties, recrystallization phenomena, occlusion, plasticity, etc.

The introduction of the necessary amount of liquid high-molecular silicon organic compound into the solid mass of dispersed substance lowers the degree of interconstituent friction, peptizes the filler particles, and facilitates their free migration. It is thus possible to achieve tighter packing.

After firing the mass obtained by the above method, the crystallization necessary for gaining the high degree of strength and uniformity which give the finished articles their high quality is achieved.

Thus, for example, the clay which is the carrier of plastic properties and the binder of the usual ceramic masses makes way for a high-molecular silicon organic compound which has a substantial effect on the quality of the product during its shaping and a subsequent firing. This is all the more important because in many cases materials containing clay are unstable and unsuitable, e.g., for the specific working conditions of the ceramic products.

By using silicon organic compounds and special chemical technology methods, substances can be obtained from which the finished product can be shaped by any method: pouring, squeezing the paste through nozzles of various shapes, powder pressing, centrifugal casting, machining the half-finished material with cutters, etc.

Our experimental investigations confirm the above theoretical position. The raw ceramic materials prepared by us, in contrast to the ordinary multi-component systems, are two-component systems, in which one phase, the silicon organic compound, is liquid and the other, a refractory metallic oxide, is solid.

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